

# TESTING OF THE DURABILITY OF LOW-CARBON CONCRETES FOR LOW AND INTERMEDIATE LEVEL NUCLEAR WASTE CONCRETE STRUCTURES

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## ABSTRACT

Extension of the Low and Intermediate Level Nuclear Waste (LILW) repositories is planned in Finland due to the existing lack of capacity to accommodate the expected volumes from decommissioning of the existing nuclear power plants. The interest of using low-carbon concrete has increased rapidly, driven by the awareness that cement manufacturing is responsible for approximately 7 % of the global carbon dioxide emissions. The objective of this research is to investigate the properties and key characteristics of low-carbon concrete with mineral additives to evaluate its suitability for use in LILW repository structures.

In this paper, five different concrete mixtures with increasing levels of Ground Granulated Blast-Furnace Slag (GGBFS) content as cement replacement were cast and subjected to tests for heat monitoring, compressive strength, modulus of elasticity, shrinkage and carbonation. The fifth mixture, containing only GGBFS slag as binder, was activated using a sodium silicate solution and dissolved sodium hydroxide, forming alkali-activated slag (AAS) composite. The findings were then compared with existing literature.

GGBFS substitution appeared to enhance the durability of concrete as it reduces its permeability. However, based on the accelerated carbonation method, the carbonation resistance emerged as a crucial factor of low-carbon concrete suitability in LILW repository environments. This research demonstrates that low-carbon concrete may possess properties different from those of ordinary Portland cement (OPC) concrete. Therefore, it is essential for supporting sustainability to thoroughly analyse and investigate their potential to ensure effective utilization in applications where they outperform OPC concrete.

Keywords: Low-carbon concrete, LILW repositories, Ground granulated blast-furnace slag, Alkali-activated slag.

## INTRODUCTION

Finland has 5 nuclear power units in operation at 2 different sites: Olkiluoto and Loviisa. The Finnish waste management policy for Low and Intermediate Level Nuclear Waste (LILW) generated by the operation and later decommissioning of NPPs foresees disposal in dedicated repositories located at the NPP sites. The repository at Olkiluoto also accommodates small radioactive waste inventories generated by Finnish healthcare, research and through industrial uses. A planned extension to the LILW facilities of both Olkiluoto and Loviisa is planned for adding the additional LILW decommissioning waste from all units (IAEA, 2020).

Considering the commissioning of new NPPs, operational and maintenance waste will be generated over time. However, an apparent larger aspect is the radioactive LILW generated during the

decommissioning of existing NPP, as the current repositories lack the capacity of accommodating the expected volumes (Becker & Mraz, 2022; Keto et al., 2022). Thus, additional repositories have been planned for this matter in both Loviisa and Olkiluoto as seen in Figure 1 (a) and (b), respectively.

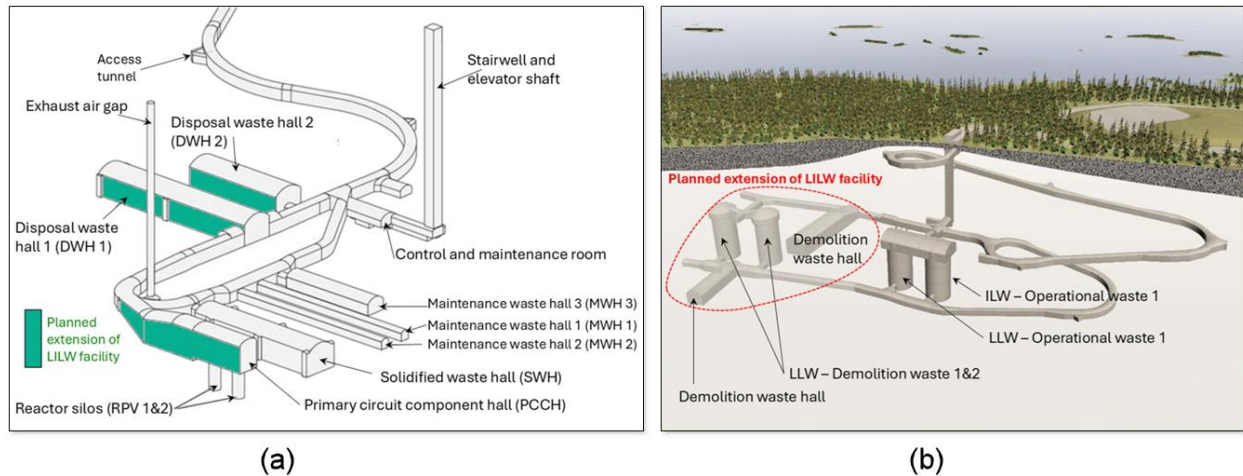


Figure 1. Operating and planned extension of (a) Loviisa and (b) Olkiluoto LILW repositories grey (Kumpula et al., 2022; Pitkääoja, 2019).

The disposal concept for low and intermediate level nuclear waste (LILW) repositories applied in Finland in rock caverns involves engineered barriers of concrete designed to remain effective for hundreds of years. These repositories, located at a depth of 60 – 110 m, are intended for the processing and disposal of waste arising from the operation of NPPs and other nuclear facilities (IAEA, 2020).

The planned extension of the LILW repositories addresses the research needs that focus on specific durable concrete mixtures with a long-term durability performance. The length of the service life of concrete at the final disposal, which can extend up to hundreds of years, should be considered in the selection of these concrete mixtures. The main objective of the research was to test durable concretes mixtures with different types of binders for the LILW repositories in Olkiluoto and Loviisa considering the enlargement of these repositories for radioactive waste generated from the decommissioning of the NPP. Low carbon concretes were proposed as a sustainable concrete type to be used for the planned extensions of the LILW repositories (Al-Neshawy & Ragaa, 2023).

The interest of using low-carbon concrete has increased rapidly, driven by the awareness that cement manufacturing is responsible for approximately 7% of the global carbon dioxide emissions (Hildebrandt et al., 2022; V. M. Malhotra, 2000). The objective of this paper is to investigate the properties and key characteristics of low-carbon concrete with mineral additives to evaluate its suitability for use in LILW repository reinforced concrete structures.

Low-carbon concrete refers to, as the name reveals, concrete that has been produced with a reduced carbon footprint compared to ordinary concrete. A commonly used measure for low-carbon concrete is the Global-Warming Potential (GWP). GWP is an index measuring the cumulative radiative forcing of a substance compared to carbon dioxide, considering its atmospheric lifetime and effectiveness in causing radiative forcing (IPCC, 2023). Thus, GWP enable the comparison of different greenhouse gasses, for instance methane and carbon dioxide which have large differences regarding initial impact and atmospheric longevity. In other words, GWP provides a standardized way to compare products with different greenhouse

gas emissions to facilitate the decisions about which emissions to prioritize to combat climate change more efficiently.

In Finland, a classification system called BY (Betoniyhdistys ry/concrete association) Low Carbon Classification has been developed for impartially categorize concrete according to their CO<sub>2</sub> emissions (Concrete Association of Finland, 2022). The method takes several aspects into account when determining the GWP. For instance, the specific emission values of the raw materials, transportation of the ingredients and energy consumption as well as the amount of waste generated during the production. Cement is known to contribute the largest amount of CO<sub>2</sub> emissions for concrete (Divsholi et al., 2014) while the specific emission for Ground Granulated Blast-Furnace Slag (GGBFS) is about 10% of cement. This is the key contributor to why cement replacement by, for instance, GGBFS significantly reduces the carbon footprint of concrete when implemented.

Low-carbon concrete emerges as a competitive choice, offering sustainability and more eco-friendly solution that not only discover an application for industrial by-products but also minimizes the environmental consequences associated with conventional concrete. Investigating the potential use of low-carbon concretes for nuclear LILW repositories is essential, as low-carbon concretes may display distinctive characteristics compared to ordinary concrete. For instance, variations in strength and durability highly influence the long-term performance of the low-carbon concrete in harsh environment, which is essential for evaluating the safety of the final disposal.

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## EXPERIMENTAL INVESTIGATION

Five different concrete mixtures with increasing levels of ground granulated blast-furnace slag (GGBFS) content as cement replacement were cast. The fifth mixture, containing only slag as binder, was activated using a sodium silicate solution and dissolved sodium hydroxide, forming alkali-activated slag (AAS) composite. Mix-design of the five concretes is presented in Table 1. The five concretes were subjected to tests for heat monitoring, compressive strength, modulus of elasticity, shrinkage, accelerated carbonation and microstructural analysis of the concrete using a petrographic method (thin-section analysis).

Table 1. Mix design of concrete mixtures (Sundström, 2024).

	Binder		w/b ratio	agg/b ratio	SP/b [%]	Activators	
	Portion [%]	Type				AA/b [%]	Type
SR-mix	100	CEM I	0,40	5	0,9	-	-
SR-SL-mix	50	CEM I	0,42	5	0,66	-	-
	50	GGBFS					
K-mix	100	CEM III/B	0,50	5	0,075	-	-
K-Si-mix	89	CEM III/B	0,44	5	0,65	-	-
	11*	Silica fume					
AAS-mix	100	GGBFS	0,48	4,8	1,05	4	WG
						6	NaOH

*b = binder, AA = alkali activator, SP = Superplasticizer, agg = aggregates and WG = Waterglass (sodium silicate).*

*\* Silica fume was designed for 10 %, because of the chemical activities of silica the actual value was 11 %.*

The calculation of low carbon classification is performed for each concrete mixture (Table 2). The emissions are calculated individually for each concrete mix based on the target values of the composition of the mix. For each raw material, the following shall be included

- Production of raw material (= quantity of raw material [kg/m<sup>3</sup>] \* specific emission [kg CO<sub>2</sub>e/kg])
- Transport of raw material to concrete station (= quantity of raw material [kg/m<sup>3</sup>] \* transport distance [km] \* specific emission [kg/CO<sub>2</sub>e/t km]))
- Energy consumption at the concrete station (electricity and heating energy) using standardised average consumption values, but emission values specific to the energy mode

Table 2. Low carbon classification of concrete mixes from BY Low Carbon Calculator (Concrete Association of Finland, 2022).

	Materials	SR-mix	SR-SL-mix	K-mix	K-Si-mix	AAS-mix
Ingredients [kg/m <sup>3</sup> ]	CEM I 42.5 N – SR3	375	189	–	–	–
	CEM III/B 42.5 L – LH/SR	–	–	358	297	–
	GGBFS (KJ400)	–	189	–	–	370
	Silica fume	–	–	–	37	–
	Aggregates	1875	1880	1791	1867	1767
	Water	151	158	178	164	179
	Superplasticizer	3,37	2,5	0,269	2,42	3,87
	Sodium hydroxide	–	–	–	–	22,2
	Sodium silicate (Water glass)	–	–	–	–	14,8
Specific emission [kg CO <sub>2</sub> e/kg]	CEM I 42.5 N – SR3	0,711	0,711	–	–	–
	CEM III/B 42.5 L – LH/SR	–	–	0,235	0,235	–
	GGBFS (KJ400)	–	0,069	–	–	0,069
	Silica fume	–	–	–	0,0053	–
	Aggregates	0,004	0,004	0,004	0,004	0,004
	Water	0,009	0,009	0,009	0,009	0,009
	Superplasticizer	1,53	1,53	1,53	1,53	1,53
	Sodium hydroxide*	–	–	–	–	1,12
	Sodium silicate (Water glass)*	–	–	–	–	1,64
Specific emissions of raw materials [kg CO <sub>2</sub> e]		281	160	93	83	89
Energy consumption [kg CO <sub>2</sub> e]**		22	22	22	22	22
Raw material transport [kg CO <sub>2</sub> e]**		21	21	21	21	21
<b>Total Specific emission [kg CO<sub>2</sub>e]</b>		<b>324</b>	<b>203</b>	<b>136</b>	<b>126</b>	<b>132</b>
Low carbon classification for C40/50 Concrete		GWP.REF	GWP.70	GWP.40	GWP.40	GWP.40
* Emission factors in kg CO <sub>2</sub> -equivalent per unit ( <a href="https://legacy.winnipeg.ca/finance/findata/matmgt/documents/2012/682-2012/682-2012_appendix_h-wstp_south_end_plant_process_selection_report/appendix%207.pdf">https://legacy.winnipeg.ca/finance/findata/matmgt/documents/2012/682-2012/682-2012_appendix_h-wstp_south_end_plant_process_selection_report/appendix%207.pdf</a> )						
** 100 km binder transportation ( <a href="https://vahahiilinenbetoni.fi/wp-content/uploads/2024/10/by_low_carbon_classification_version2_0124-2.pdf">https://vahahiilinenbetoni.fi/wp-content/uploads/2024/10/by_low_carbon_classification_version2_0124-2.pdf</a> )						

## RESULTS AND MAIN FINDINGS

### *Fresh Concrete Tests*

The fresh concrete tests (slump, density and air content) were conducted for each batch. Table 3 presents (i) the measured slump values with their corresponding slump class according to “by 65 Concrete Code 2021” (Concrete Association of Finland, 2016), (ii) the calculated fresh concrete density and (iii) measured air content.

All slump measurements could be considered as true slump. However, the flow test would be more appropriate for measuring the SR-SL-mix workability since the slump value was so high. Furthermore, the slump values measured for SR-SL-mix and K-mix batches lays on the border of slump class S4 and S3, respectively. The density of the mixtures logically decreases as the water and air content increases among them.

Table 3. Test results of fresh concrete.

Recipe	SR-mix	SR-SL-mix	K-mix	K-Si-mix	AAS-mix
Slump value, $h$ [mm]	170	220	50	120	180
Slump class	S4	S5	S2	S3	S4
Density, $D$ [kg/m <sup>3</sup> ]	2 400	2 410	2 330	2 370	2 370
Air content [%]	2,5	1,0	3,0	1,5	1,0

### *Heat Development of Concrete*

The heat release was determined according to equation (1). The air temperature during the test was approximately 20.7 °C. The fresh concrete density is presented in Table 3. The specific heat was assumed to be 800, 3800 and 900 J/(kg\*°C) for the aggregates, water and binder content, respectively. The specific heat for solid sodium hydroxide and dry mass of sodium silicate were both assumed to be 1400 J/(kg\*°C).

$$q_{con} = \frac{\rho_c c_c V_c}{m_{bnd}} \left( (T_c(t) - T_{air}) + a \int_0^t (T_c(t) - T_{air}) dt \right) \quad (1)$$

Where  $q_{con}$  is the heat accumulated in concrete [kJ/kg binder],  $\rho_c$  is the density of concrete [kg/m<sup>3</sup>],  $c_c$  is the specific heat capacity of concrete [kJ/(kg\*°C)],  $V_c$  is the volume of concrete sample [m<sup>3</sup>],  $m_{bnd}$  is the mass of the binder [kg],  $T_c(t)$  is the Measured concrete internal temperature at time  $t$  [°C],  $T_{air}$  is the measured ambient temperature [°C] and  $a$  is the Cooling factor [1/s].

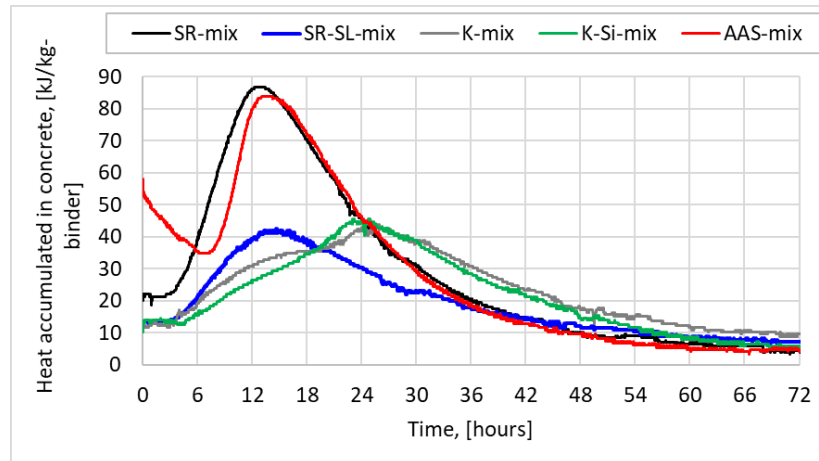


Figure 2. Heat accumulated in concrete [kJ/kg] according to (EN 12390-14, 2018).

As shown in Figure 2, it was noticed that the SR-mix and AAS-mix released heat more rapidly compared to the other mixtures. The initial temperature for the AAS-mix was also the highest due to the exothermic reaction where solid NaOH was dissolved in water before mixing started. The heat peak represents the second peak, also known as acceleration/deceleration period, and corresponds to the formation of C-S-H and C-H. For the SR-SL-, K- and K-Si-mixes containing slag, the heat release is significantly lower. For the mixtures containing CEM III/B (K- and K-Si-mix) uneven heat release was noticed. The high peak for AAS-mix (containing only slag as binder) can, of course, be explained by the alkali-activation. However, higher NaOH content has been reported to increase the intensity of the peak (Luukkonen et al., 2020).

### ***Mechanical Properties of Concrete***

The compressive strength and modulus of elasticity (MoE) values were averaged from three (3) test specimens each. Cubic compressive strength tests were conducted on 100 mm cube specimens at the 7, 28 and 91 days while the cylindrical compressive strength was determined after the MoE tests on cylindrical specimens at 28 days. Table 4 shows the obtained numerical results whereas Figure 3 graphically illustrates the cubic compressive strength.

Table 4. Averaged compressive strength and modulus of elasticity.

	Compressive strength [MPa]				Secant modulus of elasticity at 28 days [GPa]	
	Cubic			Cylindrical 28 days	Initial	Stabilized
	7 days	28 days	91 days			
SR-mix	61.3	69.6	78.2	59.4	34.8	39.1
SR-SL-mix	34.8	56.4	73.4	49.3	33.5	38.1
K-mix	30.5	40.8	44.6	37.4	32.4	36.1
K-Si-mix	35.8	48.7	53.6	43.4	33.7	37.7
AAS-mix	44.2	58.8	66.4	48.6	27.2	32.1

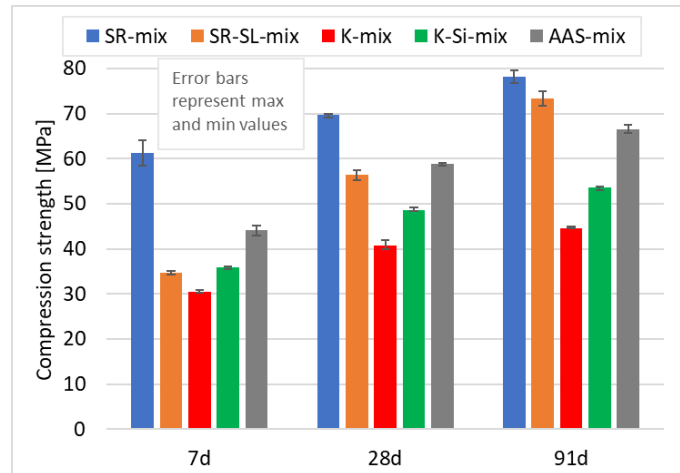


Figure 3. Average compression strength of 100mm cube specimens at 7, 28 and 91 days – according to (EN 12390-3:2001, 2001).

As shown in Figure 3, the SR-mix, containing only CEM I as binder, gained a lot of strength already after 7 days. The SR-SL-mix containing 50 % slag and 50 % cement gained slightly higher strength than the K-mix, containing CEM III/B with approximately 73 % slag. The AAS-mix has gained decent strength when compared to the other mixtures.

The modulus of elasticity MoE results show that the higher compressive strength tends to correlate with higher MoE. This trend is observed across all concrete specimens except for the AAS-mix, which exhibits significantly lower MoE than expected. Hypothetically, this is caused by differences in the microstructure of the paste, potentially due to shrinkage induced cracking.

### *Shrinkage of Concrete*

The shrinkage measurements were recorded from the second day until day 90 on two prism specimens (100\*100\*500 mm) per mixture. The averaged measurements for total shrinkage strain are presented in Figure 4.

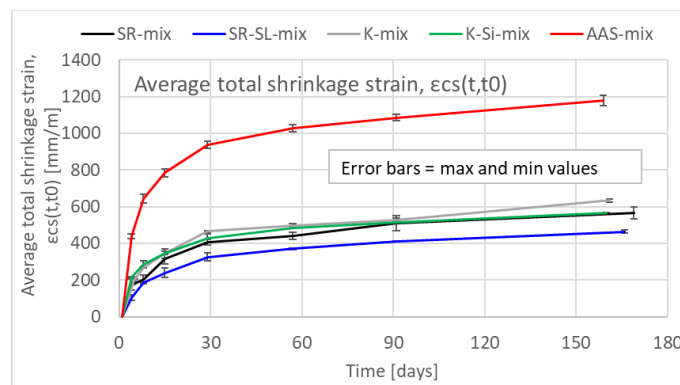


Figure 4. Mean total shrinkage strain expressed in mm/m.

It is evident that the AAS-mix specimens have shrunk significantly more than the other specimens. Additionally, its water evaporation is very low. This is reported in literature (Jiang et al., 2022; Mohamed,

2019) to be common for alkali-activated materials. High total shrinkage in alkali-activated slag pastes relates more to the development of capillary tensile forces during drying, rather than loss of moisture. The paste undergoes autogenous shrinkage, where the reaction products consume water. This property of AAS is unfavourable since the shrinkage induces cracking. The other concrete specimens exhibit similar performance to each other, comparable to that of ordinary concrete (Barr et al., 2003).

**Accelerated Carbonation Test**

The carbonation test results were obtained by measuring the carbonation depth of an approximately 50 mm wide part of a 100\*100\*500 mm prisms. The average temperature, CO<sub>2</sub> content and relative humidity in the chamber were 22.0 °C, 3.0 % and 68.7 %, respectively. The carbonation depths are expressed as the mean value of 16 measurements and illustrated in Figure 52 in accordance with the standard (EN 12390-12, 2019).

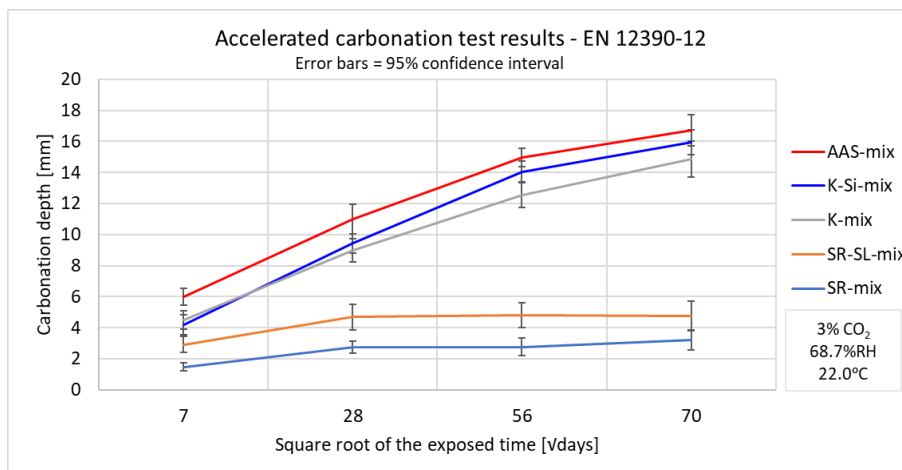


Figure 5. Measured carbonation depth based on the accelerated carbonation method.

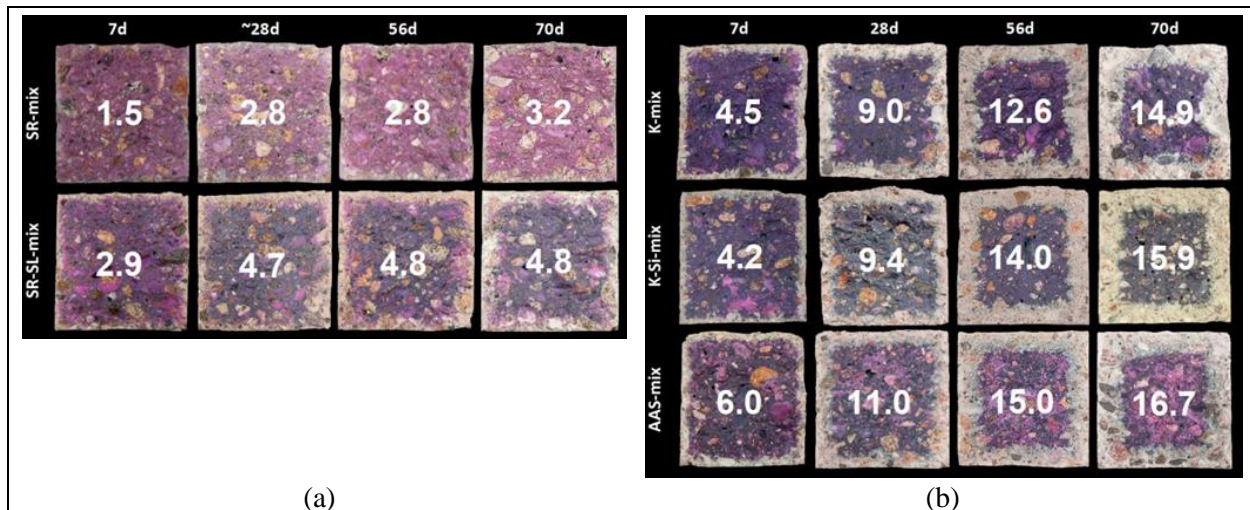


Figure 6. Accelerated carbonation depth of test specimens (a) decent carbonation resistance and (b) poor carbonation resistance.

The SR-mix and SR-SL-mix seems to have decent carbonation resistances in accelerated conditions, in comparison to K-, K-Si- and AAS-mix specimens. The standard state that the carbonation depth in concrete is expected to surpass 4 mm after 70 days of exposure. The depth for SR-mix was measured to 3.2 mm at 70d, which indicates high carbonation resistance. Additionally, SR-SL-mix have decent carbonation resistance with its measured depth of 4.8 mm at 70d. For the remaining three mixtures (K-, K-Si- and AAS-mix), the resistance to carbonation lower than the SR- and SR-SL-mixes.

***Petrographic (Thin-Section) Analysis of Concrete***

The petrographic analysis was conducted by a geotechnical consulting company in Finland. This subsection will address the most relevant findings. It is important to note that while the detected findings provide insight into the microstructure of the concrete mixtures. The most relevant finding of the analysis is the presence of microcracking in the K-, K-Si- and AAS-mix samples. Among these, the AAS-mix sample exhibited significantly stronger microcracking compared to the K-mix and K-Si-mix samples. This remarkable difference is noticeable in Figure 7 and is likely attributed to shrinkage. Additionally, in the SR-SL-mix sample, a few cracks less than 0.1 mm wide were detected between aggregates and the paste, indicating weakened adhesion.

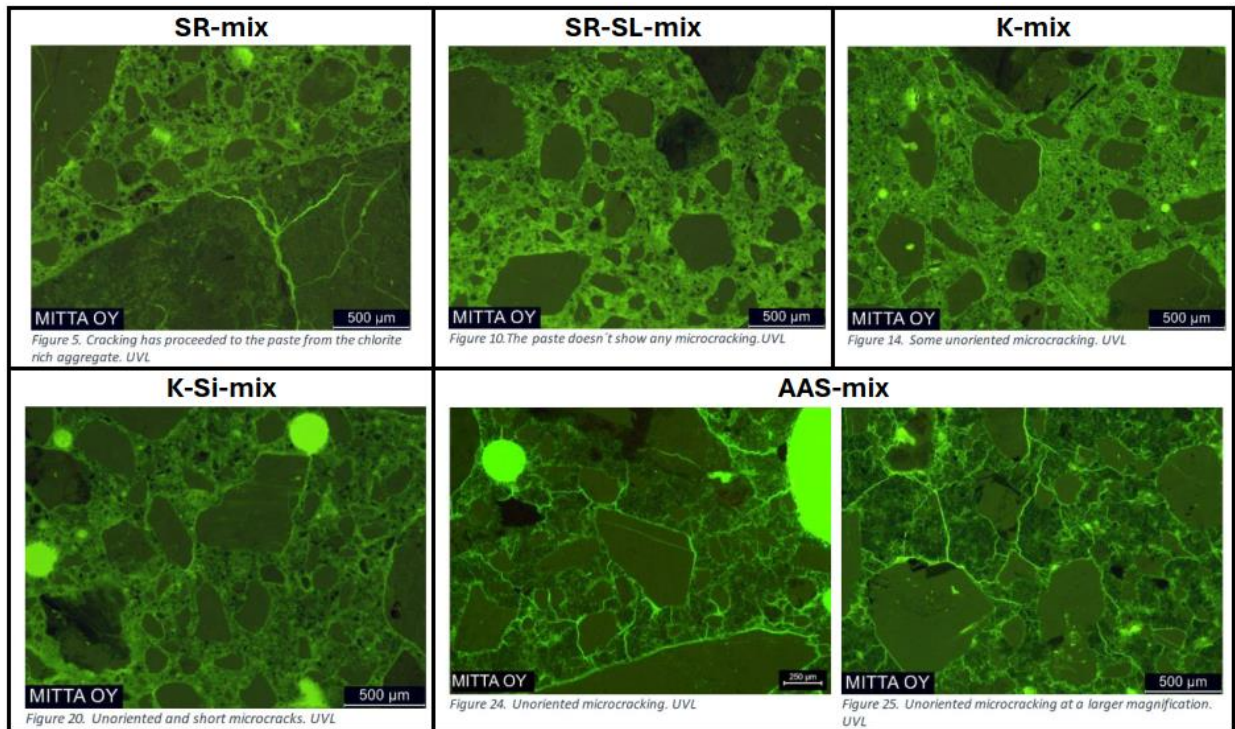


Figure 7. Crack detection on ultraviolet light figures.

The examination claimed that cracking has extended into the cement paste from one fine-grained and chloride rich aggregate. The paste crack stays very close to the aggregate and might potentially be induced by the combination of an already cracked aggregate and shrinkage of the paste. However, whether the aggregate was cracked in the first place will remain as speculations.

## CONCLUSION

With more focus than ever on embodied carbon in construction, the demand for low-carbon concrete is steadily increasing. Low-carbon concrete refers to concrete with a lower carbon footprint than ordinary concrete, achieved by substituting a portion of Portland cement with secondary cementitious materials (SCMs). These SCMs – for instance Ground Granulated Blast Furnace Slag (GGBFS) can reduce the amount of cement needed in a concrete mix, thereby lowering the carbon emissions. Another alternative to low-carbon concrete is the alkali-activated slag (AAS) composites, which are prepared from GGBFS activated through appropriate alkali solution.

The research involved casting of four different concrete mixtures with increasing levels of GGBFS content as cement replacement and one alkali-activated slag mixture, where GGBFS was activated using a sodium silicate solution and dissolved sodium hydroxide. The following durability related tests were performed on each mixture (i) monitoring of heat development in hardening concrete, (ii) compressive strength, (iii) modulus of elasticity, (iv) shrinkage and (v) accelerated carbonation depth measurements.

Based on the investigation results, the main conclusions are listed below:

- 1) Silica fume enhances the mechanical properties of low-carbon concrete obtained by cement replacement of ground granulated blast-furnace slag.
- 2) The alkali-activated slag mixed in the experimental investigation, like many other alkali-activated materials, demonstrated significant shrinkage, leading to hazardous microcracking within its paste.
- 3) Based on the accelerated carbonation method applied in the experimental investigation, the increment of cement replacement by ground granulated blast-furnace slag reduce the carbonation resistance, regardless utilizing sodium hydroxide and waterglass as activation.
- 4) In terms of carbonation resistance, the SR- and SR-SL-mix with cement replacement rates up to 50 % appears suitable for the low and intermediate level nuclear waste repository conditions.
- 5) Based on the test results, the low-carbon concrete could be utilized in applications where it outperforms ordinary Portland cement.

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